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Assessment of phosphorus loading dynamics in a tropical reservoir with high seasonal water level changes

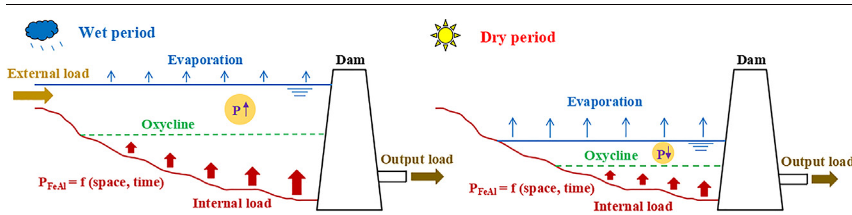
Iran E. Lima Neto [a,*], Pedro H.A. Medeiros [b], Alexandre C. Costa [c], Mario C. Wiegand [a], Antônio Ricardo M. Barros [a], Mário U.G. Barros [d]

[a] Department of Hydraulic and Environmental Engineering, Federal University of Ceará, bl. 713, 60.451-970 Fortaleza, Brazil
[b] Federal Institute of Education, Science and Technology, Campus Fortaleza, Av. Treze de Maio, 2081, 60.040-531 Fortaleza, Brazil
[c] Institute of Engineering and Sustainable Development, University of International Integration of the Afro-Brazilian Lusophony, 62.790-970 Redenção, Brazil
[d] Water Resources Company of the State of Ceará COGERH, Rua Adualdo Batista, 1550, 60.824.140 Fortaleza, Brazil

HIGHLIGHTS

- P linked to iron and aluminum (P_{FeAl}) increased over time and along the reservoir bed.
- Internal P loads were estimated from P_{FeAl} and variable heights of the anoxic layer.
- Adaptation of Vollenweider's model improved P predictions in the water.
- Internal load prevailed in the wet period and was slightly lower than external load.
- The internal load would increase by 6-fold if the reservoir were 70 years older.

GRAPHICAL ABSTRACT



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ABSTRACT

Nutrient accumulation in man-made reservoirs has been documented worldwide. Therefore, quantifying phosphorus loading and understanding its temporal dynamics in reservoirs is mandatory for sustainable water management. In this study, the Vollenweider's complete-mix phosphorus budget model was adapted to account for high water level variations, which are a common feature in tropical reservoirs, and for internal phosphorus loads. First- and zero-order kinetics were adopted to simulate phosphorus settling and release from the sediment layer, respectively, considering variable area of phosphorus release according to the height of the anoxic layer. The modeling approach was applied for a 52-months period to a 31-years-old reservoir located in the semiarid region of Brazil with 7.7 hm^3 storage capacity. The simulations were supported by hydrological, meteorological and water quality data, as well as analyses of phosphorus partitioning of the reservoir bed sediment. The external phosphorus load was estimated from a relationship adjusted between inflow and phosphorus concentration, revealing an u-shaped pattern. Sedimentary phosphorus linked to iron and aluminum (P_{FeAl}) increased over time and along the reservoir. Such measurements were used to estimate the internal phosphorus load, i.e., the yield from the bed sediments to the water column. The adaptations proposed to the model's structure improved its capacity to simulate phosphorus concentration in the water column, from "not satisfactory" to "good". We estimate that the internal phosphorus load currently accounts for 44% of the total load. It prevailed during the wet period, when reservoir stratification and hypolimnetic hypoxia were more notable, resulting in higher phosphorus concentration in the water column due to the combined effects of internal and external loadings. However, if the reservoir were 70 years older, the internal load would reach 83% of the total and the reservoir would become a source instead of a sink of phosphorus.

* Corresponding author.
E-mail addresses: iran@deha.ufc.br (I.E. Lima Neto), phamedeiros@ifce.edu.br (P.H.A. Medeiros), cunhacos@unilab.edu.br (A.C. Costa), mario.barros@cogerh.com.br (M.U.G. Barros).

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1. Introduction

The quantification and reduction of phosphorus (P) loading has been one of the main challenges in eutrophication control of inland waters (Rattan et al., 2017; Le Moal et al., 2019). The total P loading is normally divided into two main sources: external and internal loadings. P input from streams is called external P loading, which may be further divided into point and nonpoint sources (Bowes et al., 2008). On the other hand, P released from lake sediments is known as internal loading (Nikolai and Dzialowski, 2014). High phosphorus concentrations in the sediments are a result of a long-term accumulation process from P entering and settling at the lake bed. The challenge, though, is distinguishing between the internal and external loadings, in order to optimize the reduction of input loads and assure the reservoirs' sustainability (Nürnberg et al., 2013).

The external P loading usually represents a higher percentage in an annual P budget and its relevance has formed the basis for classic predictive models such as the Vollenweider's P model (Chapra, 2008). A major fraction of P entering a lake ecosystem, however, is usually stored in the bottom sediment (Kiani et al., 2020), which might return to the water column under appropriate conditions (Moura et al., 2020). Phosphorus releasing from sediments results from physical and/or biogeochemical mechanisms that operate on different temporal and spatial scales in a complex manner. For instance, in shallow lakes, phosphorus resuspension due to the effect of wind shear is a potential source of internal loading (Søndergaard et al., 2003; Araújo et al., 2019; Mesquita et al., 2020). On the other hand, in deep lakes, more prone to stratification of the water column and hypoxia of the deeper water layers, P release from anoxic sediments is a common source of internal P loading (Chapra and Canale, 1991; Wang et al., 2016; Tu et al., 2019). Therefore, internal loading might pose a high risk of becoming an additional relevant P source, contributing to the total lake P bioavailability and, consequently, to the deterioration of water quality (Doan et al., 2018).

The quantification of external P loading is relatively straightforward by measuring flow rate and P concentration at the lake inlets (Bowes et al., 2008) or by performing mass balance studies (Rocha and Lima Neto, 2021a). In contrast, the quantification of internal P loading is still challenging, which depends not only on lake hydrodynamics, but also on water quality and sediment composition. Several ways to estimate the internal P loading include: determination from hypolimnetic P increases, regression analysis, time-dynamic modeling, estimates from anoxic active area and mass balance approaches (Nürnberg, 2009; Nürnberg et al., 2012; Rocha and Lima Neto, 2021b). Previous research has demonstrated that specific sedimentary phosphorus fractions, especially P linked to iron and aluminum (P_{FeAl}), are responsible for promoting the internal loading from anoxic sediments (Rydin, 2000; Wang et al., 2006; Ribeiro et al., 2008; Ding et al., 2016; Tu et al., 2019; Kiani et al., 2020). Recent laboratory studies conducted by Moura et al. (2020) have shown that the release of P from anoxic sediments is directly related to the concentration of P_{FeAl}, which in turn is related to reservoir age. However, a conceptual model based on P fractions and variable anoxic active areas, which are expected to impact P dynamics in real lakes and reservoirs, particularly those subject to significant variations in water level, has not been found in the literature.

For tropical regions, most water supply reservoirs are recurrently eutrophic or hypereutrophic, which makes the P pollution issue a great concern for integrated water resources management (Lima et al., 2018; Lacerda et al., 2018; Lira et al., 2020; Raulino et al., 2021; Rocha and Lima Neto, 2021a, 2021b; Wiegand et al., 2021). The high seasonal and interannual variation of reservoir water level in these regions potentially impacts lake stratification and oxygenation patterns, as well as P dynamics, accounting for both the internal and external P loadings. The dynamics of external P loading in tropical reservoirs has been recently studied by Rocha and Lima Neto (2021a). On the other hand, the seasonal variation of total P loading has been investigated by Rocha and Lima Neto (2021b). Nevertheless, to the authors' knowledge, the dynamics of internal P loading in tropical reservoirs, where water level variation is a major issue, has not been reported in the literature.

The main objective of this work was to innovatively assess phosphorus loading dynamics in a reservoir with high seasonal water level changes in the Brazilian semiarid region. The specific objectives were: (i) to obtain a relationship to describe the external load entering the reservoir; (ii) to investigate the spatio-temporal variation of sedimentary P fractions in the reservoir; (iii) to investigate the variability of the heights of the anoxic layer; (iv) to propose a novel model to describe the internal load based on P fractions and variable heights of the anoxic layer; (v) to model P dynamics considering both the internal and external loadings assessing the contribution of those sources to P concentrations in the water column; (vi) to investigate the impact of reservoir age on P budget. The results from this study improve the knowledge of limnology of tropical reservoirs and the proposed model is a potential tool for integrated water resources management.

2. Material and methods

2.1. Study area: hydrology and total phosphorus sources

This study was conducted in São José I, a 31-years-old reservoir located in the semiarid northeast of Brazil with storage capacity of 7.7 hm^3, maximum height of 10 m and maximum surface area of 2.15 km^2 (Fig. 1). Annual rainfall and potential evaporation in the region are roughly 700 mm and 2500 mm, respectively, with rainfall concentrated from January to May, accounting for 83% of the annual amount. Such pattern results in a strong seasonal variation of the reservoir water level: streamflow events may occur over a period of up to about four months, and the inflow to the reservoir persists certainly null over the rest of the year (Costa et al., 2021), when the water level is significantly reduced due to the high evaporation rates. Interannual variability is also quite high (Zhang et al., 2021), with the coefficient of variation of annual streamflow reaching up to 1.4, leading to extreme events, such as hydrological droughts and floods (Costa et al., 2021; Van Langen et al., 2021).

According to Ceará (2021), the catchment area of São José I reservoir is 15.5 km^2, which is characterized by mild slopes (0–15°). The soils are relatively shallow as they are located in the crystalline geological formation and the most representative soil types are the red-yellow argisol, litolic neosol and chromic luvisol. These conditions together with the tropical semiarid climate favor the existence of intermittent rivers and practically no baseflow. In fact, regional groundwater flow with potential impacts on reservoir water quality only occurs on the borders of the State of Ceará and in a large sedimentary basin in the Southeast, as reported by Wiegand et al. (2021). Hence, since the São José I reservoir is located in the central part of the State of Ceará (see Fig. 1), which is characterized by the crystalline bedrock with shallow overlying soils and low hydraulic conductivity and porosity, groundwater interaction was neglected. Moreover, as the studied reservoir is in a semiarid zone, the vegetation along its shore areas is very sparse and the effect of evapotranspiration on the reservoir water budget was also neglected. Such simplifications are supported by several studies carried out in the Brazilian semiarid region (Campos et al., 2016; Raulino et al., 2021; Rabelo et al., 2021).

The catchment of São José I reservoir is located in the Banabuiú River basin, which presents the following distribution of P pollution sources (Rocha and Lima Neto, 2021a): soil (45%), agriculture (30%), sewer (18%) and livestock (7%). Therefore, it is expected that the seasonal inflows to the reservoir will carry the phosphorus produced from these sources to generate the external P load and, consequently, the internal P load.

2.2. Field and laboratory studies

The assessment of phosphorus loads comprised a period of over four years (Jan 2017 - Apr 2021), and was based on measured data and modeling. Daily hydrological data (rainfall and reservoir water level) from the monitoring program of the Water Resources Company of the State of Ceará (COGERH), as well as monthly averages of potential evaporation provided by the Brazilian National Institute of Meteorology (INMET), were

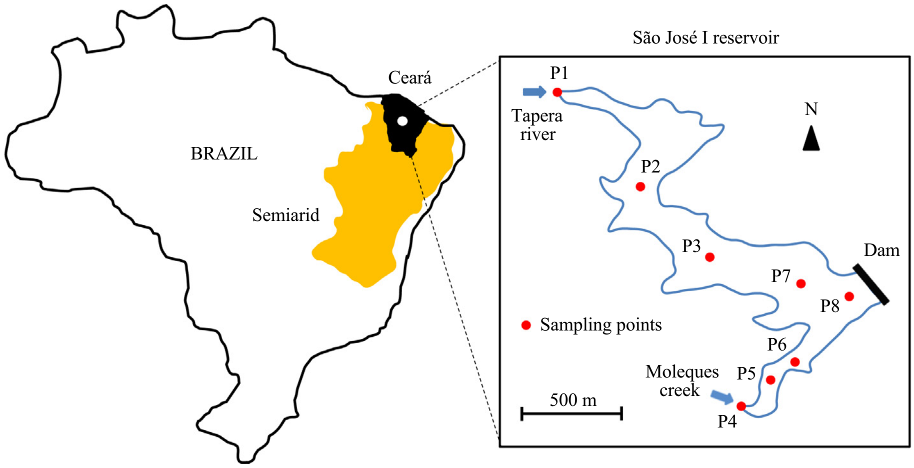

Fig. 1. Study area map: location of the State of Ceará and semiarid region in Brazil (left panel) and São José I reservoir with indication of sampling points (right panel).

used to simulate the reservoir water storage dynamics. The curves relating water level, area and volume of the reservoir were also provided by COGERH.

As sketched in Fig. 1, the sampling sites covered the two branches of the reservoir: the first one including points P1, P2, P3, P7 and P8, and the second one including points P4, P5, P6 and P8.

Water quality data at point P7 was provided by COGERH, which carried out 16 campaigns during the study period (3 to 4 campaigns per year), both in the wet and dry seasons. The laboratory analyses were conducted according to APHA (2005) and the following parameters were assessed: total phosphorus, total nitrogen, chlorophyll-a and transparency (Secchi Disk). Additionally, a water quality probe (YSI 6600 V2) was used to quantify the depth variation of water temperature, dissolved oxygen (DO) and pH, which were used to identify stratification patterns and the critical level of DO for phosphorus release from the bottom sediment to the water column, i.e., the occurrence of internal P load. Wind speed at point P7 was also measured with an anemometer by COGERH, this data was used to interpret phosphorus resuspension from bottom sediments.

To complement the hydrological and water quality data provided by the Ceará State institutions, we conducted the following field/laboratory work:

- Four field measurements of streamflow and total phosphorus concentration in the Tapera river (point P1), immediately upstream the São José I reservoir, including one survey in December 2018 and three in March, April and May 2019. The streamflows were measured by using an electromagnetic propeller flow meter MiniWater20, with speed range of 0.02 to 5.0 m/s, as described by Mesquita et al. (2020). The analyses of total phosphorus concentration were carried out in the Environmental Sanitation Laboratory of the Federal University of Ceará (LABOSAN), according to APHA (2005). Monitoring of the Moleques creek (point P4) was not possible as it remained dry during the campaigns;

- Two sediment sampling campaigns at the reservoir bed (points P1 - P8) in October 2018 and October 2019, for analyses of phosphorus partitioning in LABOSAN. Sediment sampling was performed in triplicate up to a depth of 5 cm, which is considered to be the depth of sediment containing the potentially available phosphorus for release under anoxic conditions (Steinman et al., 2007; Tu et al., 2019). The analysis of P fractions in the sediment was carried out following the same methodology described by Moura et al. (2020): total phosphorus (P_T) content was analyzed as soluble phosphate after extraction by heating the sediment at 500 °C for 1 h, followed by high temperature (340 °C) acid hydrolysis, according to Hedley et al. (1982). P_T, in turn, can be subdivided into inorganic (P_I) and organic phosphorus (P_O). P_O was obtained by subtracting P_T from P_I concentration [Eq. (1)]. Extraction of P_I from the sediment was carried out by sequential chemical fractionation until that fraction became soluble phosphate. This method consists of sequentially fractioning P_I into mobile phosphorus (P_M), iron and aluminum-bound phosphorus (P_{FeAL}), calcium-bound phosphorus (P_{Ca}) and residual phosphorus (P_{Re}), as described by Rydin (2000). For the analysis of soluble phosphate, the method 4110-B from APHA (2017) was used. Note that the above-mentioned analysis of P fractions in the sediment is necessary to estimate the internal load, according to Moura et al. (2020).

$$P_O = P_T-(P_M + P_{FeAl} + P_{Ca} + P_{Re}) \quad (1)$$

2.3. Hydrological modeling

Although daily reservoir water level was available, which enables the estimation of inflows to the reservoir from a water balance calculation, we performed a runoff simulation to compute streamflow. Such procedure reduces the uncertainties during high flows, when the reservoir is overspilling and the temporal resolution (daily) of the measured data does not take into account the intra-daily water level variation and, therefore, prevents estimating the spillage and the inflow with high accuracy. Runoff was simulated daily with the parsimonious SCS/CN method and using the rainfall time series available at the Brazilian National Water Resources Information System, from the National Water and Sanitation Agency (ANA). The model parameter (CN) was calibrated to match the average runoff coefficient reported for the region by the Ceará State Foundation of Meteorology and Water Resources (FUNCEME).

For validation of the runoff estimations, water balance was computed for the São José I reservoir according to Eqs. (2) to (4):

$$dV/dt = Q_{in}(t)-Q_{out}(t) \quad (2)$$

$$Q_{in} = Q_R + Q_P + Q_G \quad (3)$$

$$Q_{out} = Q_E + Q_I + Q_S + Q_W \quad (4)$$

where: dV/dt ($m^3\ day^{-1}$) is the time variation of the stored volume V; Q_{in} represents the inflows (runoff - Q_R, direct precipitation on the lake - Q_P; underground recharge - Q_G); and Q_{out} represents the outflows (evaporation - Q_E, infiltration - Q_I, spillage - Q_S, withdrawal for water supply - Q_W), all flows being calculated in $m^3\ day^{-1}$. As already discussed in Section 2.1, the groundwater interaction terms (Q_G and Q_I) were neglected in this study. For details on the reservoir water balance calculation, please see Brasil and Medeiros (2020).

2.4. Total phosphorus modeling

In this study, we adapted the original complete-mix model of Vollenweider (see Chapra, 2008) in order to describe the phosphorus budget in the water accounting for high water level variations in the reservoir and the internal P loading (Eq. (5)):

$$d(V.P_{T,out})/dt = Q_R.P_{T,in} - (Q_S + Q_W).P_{T,out} - k_s.V.P_{T,out} + k_r.A_{sed} \quad (5)$$

where: $P_{T,in}$ is the total phosphorus concentration in the runoff entering the reservoir (mg m^{-3}); $P_{T,out}$ is the total phosphorus concentration in the water column assuming complete-mix (mg m^{-3}); k_s is the first-order settling rate of total phosphorus in the water column (month^{-1}); k_r is the zero-order release rate of total phosphorus from the sediment to the water column (mg m^{-2} month^{-1}); and A_{sed} is the surface area of the sediment zone where total phosphorus is released to the water under anoxic conditions (m^2). Note that the models describing the settling and release rates were obtained from Rocha and Lima Neto (2021a, 2021b) and Moura et al. (2020), respectively.

The assumption of complete-mix is consistent with the data reported for Brazilian semiarid reservoirs. For instance, Lima (2016) measured P_T at five different points distributed over a 30 hm^3 reservoir and obtained a coefficient of variation (CV) < 20%, while Andrade et al. (2020) performed measurements of P_T at seven different points in a 1940 hm^3 reservoir and obtained CV < 50%. On the other hand, the temporal variation of P_T, including the wet and dry seasons, was much higher (CV of up to about 200%). This suggests that taking the deepest station (point P7) as a reference of P_T for the studied reservoir, which is relatively small (7.7 hm^3), is a reasonable approximation that is consistent with the complete-mix assumption.

Observe that Eq. (5) accounts for the effects of P accumulation due to volume/concentration variation [$d(V.P_{T,out})/dt$], external P loads from runoff [$Q_R.P_{T,in}$], output P load due to spillage and withdrawal [$-(Q_S + Q_W).P_{T,out}$], P decay due to settling [$-k_s.V.P_{T,out}$], and P release from sediments or internal load [$k_r.A_{sed}$]. While the reservoir water balance (see Eqs. (2)–(4)) is solved for the entire times series (Jan 2017 - Apr 2021) and the accumulation and output P load were obtained directly from measurements, the other terms (external/internal loads and settling) were calculated based on the studies of Moura et al. (2020) and Rocha and Lima Neto (2021a, 2021b), as described in the Results and Discussion section. Hence, Eq. (5) can be solved numerically by using a simple explicit finite difference scheme (see Section 3.3 in supplementary material) in order to predict the values of $P_{T,out}$ based on previous month values of the other variables in the equation. Table S2 in supplementary material shows all model parameters and their respective values.

To assess the improvement promoted by the modifications to the original phosphorus budget model, three modeling schemes were tested: i) Settling model, which consists of Eq. (5) without the release term [$k_r.A_{sed}$]; ii) Settling and release model with fixed area (A_{sed}), adopting Eq. (5) with a fixed value for the area of sediment where total phosphorus is released to the water; and iii) Settling and release model with variable area (A_{sed}), which is the same as ii but considering that, as the levels of the water surface and the anoxic layer change, the area where phosphorus is released changes accordingly. Note that the adaptations of this model to the original Vollenweider's version include the variability of water level (or reservoir volume), the addition of the internal load dynamics by using a novel formulation, and a different parameterization for the settling process, as detailed in the supplementary material (see Section 3). This model also differs from the classical one proposed by Chapra and Canale (1991), in which two P budget equations were used, one for the water column and the other for the bed sediment. Additionally, Chapra and Canale's model also assumed constant V and A_{sed}, as well as first order reaction for the release rate, which made the model dependent on P concentration in the sediment. Moreover, constant velocities were considered to describe the settling and release rates, differently from the model proposed in the present study.

The external/internal loads and settling terms could not be obtained directly from measurements, and were calculated as following:

i) The external P load for each month was calculated from the product of the computed inflow (Q_R) by the total phosphorus concentration at the reservoir inlet ($P_{T,in}$), which was obtained from the correlation shown in the results section, Fig. 2: $P_{T,in} = 41Q_R^{-0.85} + 42Q_R^{1.0}$ ($R^2 = 0.99$). This method has been validated by Rocha and Lima Neto (2021a) for reservoirs located in the Brazilian semiarid region;

ii) The internal P load was assumed to occur when the DO level in the hypolimnion was lower than 1.5 mg.L^{-1} (Chapra and Canale, 1991; Wang et al., 2016; Moura et al., 2020). Thus, the surface areas of the sediment zone (A_{sed}) where total phosphorus is released to the water were obtained from the variable heights of the anoxic layer (h_a = 5–10 m) (see Fig. 5 in the results section) and the water level-area curve provided by COGERH, which has the deepest station (point P7) as a reference. Since low hypolimnetic DO levels (hypoxia) are related to thermal stratification of the water column, we assumed that the elevation of the thermocline/oxycline was the same inside the lake, as observed by the authors in the Castanhão reservoir, the largest in the Brazilian semiarid region (on-going research). This means that the anoxic layer at different points within the lake will have heights varying from zero to h_a. Observe that the same procedure but considering a fixed height of the anoxic layer was adopted by Chapra and Canale (1991). For each measurement (usually taken at the end of a particular month), the value of h_a was assumed constant for the present and subsequent months. The adopted anoxic factor (AF = 60 days), which represents the number of days that sediment area is overlain by anoxic water, is consistent with the results obtained from the correlation of Nürnberg (1995) as a function of total phosphorus concentration, water depth and lake surface area (see Fig. S3 in supplementary material). This correlation has already been validated to lakes world-wide including arid environments (Townsend, 1999; Kiani et al., 2020). On the other hand, the zero-order release rate of total phosphorus from the sediment to the water (k_r) was obtained from the correlation proposed by Moura et al. (2020), in which k_r is a function of P_{FeAl}: $k_r = 0.0196P_{FeAl}$. Notice that this correlation was obtained for three reservoirs located in the same basin (Banabuiú River basin) of São José I reservoir. The application of this correlation together with the data in Fig. 3 resulted in an average value of k_r = 165.0 mg.m^{-2}.month^{-1}) for current conditions. The same procedure was adopted to simulate the phosphorus budget for a hypothetical condition of reservoir age of 100 years, leading to k_r = 999.6 mg.m^{-2}.month^{-1}, but keeping the hydrological and external load regimes the same for current conditions. Finally, the internal load for each month was calculated by multiplying the spatial averaged value of k_r by the corresponding area of the sediment zone (A_{sed});

iii) The first-order settling rate k_s was obtained from the correlation given by Rocha and Lima Neto (2021a, 2021b): $k_s = 4/(RT)^{0.5}$, in which RT is the average water residence time in the reservoir. This resulted in k_s = 0.34 month^{-1}.

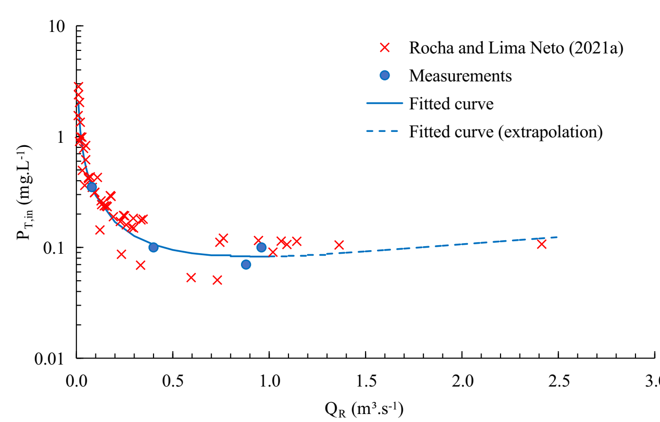

Fig. 2. Reservoir inflow versus total phosphorus concentration obtained from field measurements and data available for similar reservoirs in the State of Ceará. Measurements taken at P1 in December 2018 and March, April and May 2019.

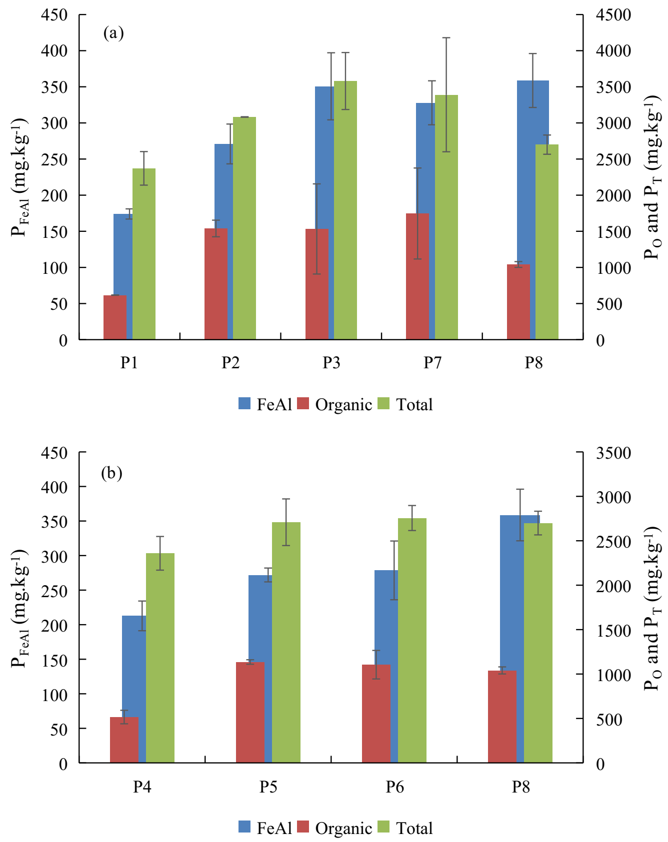

Fig. 3. Spatial variation of sedimentary phosphorus linked to Fe and Al (P_{FeAl}) (primary axis), as well as organic phosphorus (P_O) and total phosphorus (P_T) (secondary axis): (a) First branch of the reservoir, including points P1, P2, P3, P7 and P8; and (b) Second branch of the reservoir, including points P4, P5, P6 and P8. Error bars indicate standard deviations. Measurements taken in October 2018 and October 2019.

2.5. Analysis of model performance

The performance of the hydrological and total phosphorus modeling was assessed with the Nash and Sutcliffe (1970) coefficient:

$$NSE = 1 - \left[Sum(Y_{meas,i} - Y_{mod,i})^2 / Sum(Y_{meas,i} - Y_{med})^2\right] \quad (6)$$

where: $Y_{meas,i}$ is the variable value measured at time step i; $Y_{mod,i}$ is the variable value modeled at time step i; Y_{med} is the average measured value of the variable during the simulation period. NSE varies from - inf. to 1, and the closer its value is to 1, the better is the model performance. NSE values lower than 0 indicate that the predictive capacity of the model is less than simply adopting the average measured value. The ratio of root mean square error and standard deviation of measured data (RSR) was also used to assess the performance of the total phosphorus model schemes, with RSR values close to zero showing very good model performance (see Moriasi et al., 2007, 2015).

2.6. Total phosphorus budget analysis

Finally, the total phosphorus budget including the individual contributions of external load ($Q_R.P_{T,in}$), settling rate ($-k_s.V.P_{T,out}$), release rate ($k_r.A_{sed}$) and output load [$-(Q_S + Q_W).P_{T,out}$], for both the 31-years-old and 100-years-old reservoir conditions, was performed in order to evaluate the importance of each of these P transport process and the potential changes from one reservoir age condition to another.

3. Results and discussion

3.1. Hydrological analysis

High seasonal variation of water volume V stored in the São José I reservoir (from about 1 to 7.7 hm^3, which corresponded to water level h changes from about 4 to 10 m), was observed as a result of inflows occurring predominantly from January to May, while evaporation losses dominate from June to December (see Fig. S1 in supplementary material). Similar behaviors have been reported for other reservoirs located in the Brazilian semiarid region (for instance, by Campos et al., 2016; Lira et al., 2020; Zhang et al., 2021). Fig. S1 also shows the volumes modeled according to the above-mentioned hydrological scheme, resulting in a Nash-Sutcliffe coefficient (NSE) of 0.86, which can be considered as "very good", according to Moriasi et al. (2007). The computed inflows (Q_R), estimated with the calibrated SCS/CN model, ranged from zero to 3.6 m^3/s, and were used later together with the total phosphorus concentrations ($P_{T,in}$) at the reservoir main inlet (point P1, see Fig. 1) in order to estimate the external loading, as described by Rocha and Lima Neto (2021a). Additionally, Fig. S2 (supplementary material) shows the resulting reservoir water budget in percentage of inflow over the study period: evaporation (34%), withdrawal (22%), spillage (47%) and volume variation (−3%). This confirms that due to high evaporation rates, the total reservoir inflow is higher than the outflows through withdrawal and spillage, which potentially promotes phosphorus retention in the reservoir, as observed by Rocha and Lima Neto (2021b) for other 18 reservoirs in the Brazilian semiarid region.

3.2. Relationship between inflow and total phosphorus concentration

Fig. 2 shows a regression adjusted between reservoir inflow (Q_R) and total phosphorus concentration measured at point P1: $P_{T,in} = 41Q_R^{-0.85} + 42Q_R^{1.0}$ ($R^2 = 0.99$), which is consistent with the data reported by Rocha and Lima Neto (2021a) for other reservoirs located in the State of Ceará with similar capacities (up to about 20 hm^3) ($R^2 = 0.93$). The observed u-shaped pattern of the curve is expected for non-point source dominated catchments, as in the present case, where 75% of P pollution sources originate from soil and agriculture (see Section 2.1). It is important to mention that physical-based approaches to estimate nutrient mass fluxes at the catchment scale are of difficult implementation in data-scarce regions, such as the Brazilian northeast, and empirical streamflow-concentration curves may be an alternative with acceptable precision (Bowes et al., 2008; Rattan et al., 2017). Hence, the computed inflows Q_R (see Section 3.1) were used to estimate the total phosphorus concentrations at the reservoir inlet by using the fitted inflow-concentration curve ($P_{T,in} = 41Q_R^{-0.85} + 42Q_R^{1.0}$), and then estimate the external loading on a monthly basis by the product of Q_R and $P_{T,in}$.

3.3. Sedimentary phosphorus analysis

The results of phosphorus partitioning for the sediment samples collected at points P1-P8 for the 2018 and 2019 campaigns are shown in Table S1 (supplementary material). The coefficient of variation of each fraction, as sediment sampling was performed in triplicate, ranged from 0.6 to 16.2%, which can be considered relatively small for the purpose of the present study. Consistently with Wang and Liang (2015), Tu et al. (2019) and Thin et al. (2020), phosphorus in the sediment was mainly present as the inorganic form ($P_I = P_M + P_{FeAl} + P_{Ca} + P_{Re}$). Moreover, the values for each phosphorus fraction are within the ranges reported in the literature for reservoirs located in the Brazilian semiarid region (Moura et al., 2020; Cavalcante et al., 2018, 2021) and in other climate regions including subtropical and temperate zones (Rydin, 2000; Wang et al., 2006; Tu et al., 2019; Thin et al., 2020).

Fig. 3 shows the spatial variation of sedimentary P linked to Fe and Al (P_{FeAl}), according to the two branches of the reservoir: the first one including points P1, P2, P3, P7 and P8, and the second one including points P4, P5, P6 and P8. A clear trend can be observed: P_{FeAl} tends to increase from riverine to lacustrine zones. This can be attributed to the reduced flow velocity along the reservoir which results in a progressive accumulation of P_{FeAl} at the lake sediment bed. The reduced flow velocity favors P settling from riverine to lacustrine zones, consistently with the results obtained by Steinman et al. (2007) and Wang et al. (2016). On the other hand, the spatial correlation with the other inorganic fractions of P shown in Table S1 (supplementary material) was low ($R^2 < 0.1$), while the correlations with the organic fraction (P_O) and total phosphorus (P_T) were relatively high ($R^2 > 0.5$), as depicted in Fig. 3. This corroborates the results of Wu et al. (2021), in which the content of Fe-bound phosphorus and P_O increase due to the deposition of suspended matter, thus increasing P_T.

The temporal variation of P_{FeAl} can be verified in Fig. 4, which shows a comparison of P_{FeAl} in the lacustrine zone of reservoirs with different ages located in the Brazilian semiarid region, obtained by using similar phosphorus partitioning schemes (Moura et al., 2020; Cavalcante et al., 2018, 2021). Note that in our case, the average value between points P7 and P8 (assumed in the lacustrine zone) was taken for comparison purposes. The results from this study and Cavalcante et al. (2021) confirm the high correlation between reservoir age and P_{FeAl} proposed by Moura et al. (2020) and the progressive accumulation of P_{FeAl} in the Brazilian semiarid reservoirs, as a consequence of inexistent control measures such as load reduction, as discussed by Welch and Cooke (2005). Note that an overall P_T mass balance between the reservoir inlet and outlet [$Q_R.P_{T,in}$ - (Q_S + Q_W).$P_{T,out}$] resulted in a retention rate of approximately 45% of the total external load, which suggests that sedimentary P_T (as well as P_O and P_{FeAl}, see Fig. 3) increases over time. This net retention rate is within the typical range of 40–90% reported by Hejzlar et al. (2006) and Rocha and Lima Neto (2021b) for reservoirs located in temperate/tropical regions. P_{FeAl} enrichment in sediments according to reservoir age is particularly important for water management: in their laboratory-scale experiment, Moura et al. (2020) demonstrated that this fraction can be released from the sediment layer under anoxic conditions, contributing to the increase of P concentration in the water.

3.4. Vertical profiles of temperature and dissolved oxygen

Fig. 5 shows vertical profiles of water temperature (T) and dissolved oxygen (DO) concentration measured at point P7 from the water surface, which is highly variable over time. As expected, low hypolimnetic DO levels (hypoxia) were related to thermal stratification of the water column. Thus, a critical DO level for P release of 1.5 mg.L^{-1} was assumed, following Chapra and Canale (1991), Wang et al. (2016) and Moura et al. (2020). Note that here we neglected the effect of pH on P release, as pH was always within a narrow range of 6–8, which is typical of Brazilian semiarid rivers and reservoirs (Molisani et al., 2010; Freire et al., 2021) and unlikely to favor sediment P release (Tu et al., 2019). On the other hand, wind speed was under relatively calm conditions (<5 m/s), suggesting that sediment/phosphorus resuspension was negligible, even in the shallow parts of the lake (Bengtsson and Hellström, 1992; Madsen et al., 2001; Araújo et al., 2019).

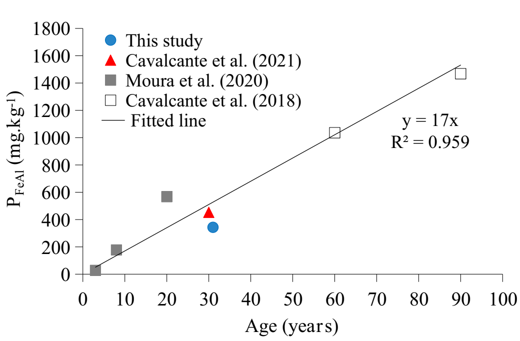

Fig. 4. Temporal variation of sedimentary phosphorus linked to Fe and Al (P_{FeAl}) for reservoirs located at the Brazilian semiarid region.

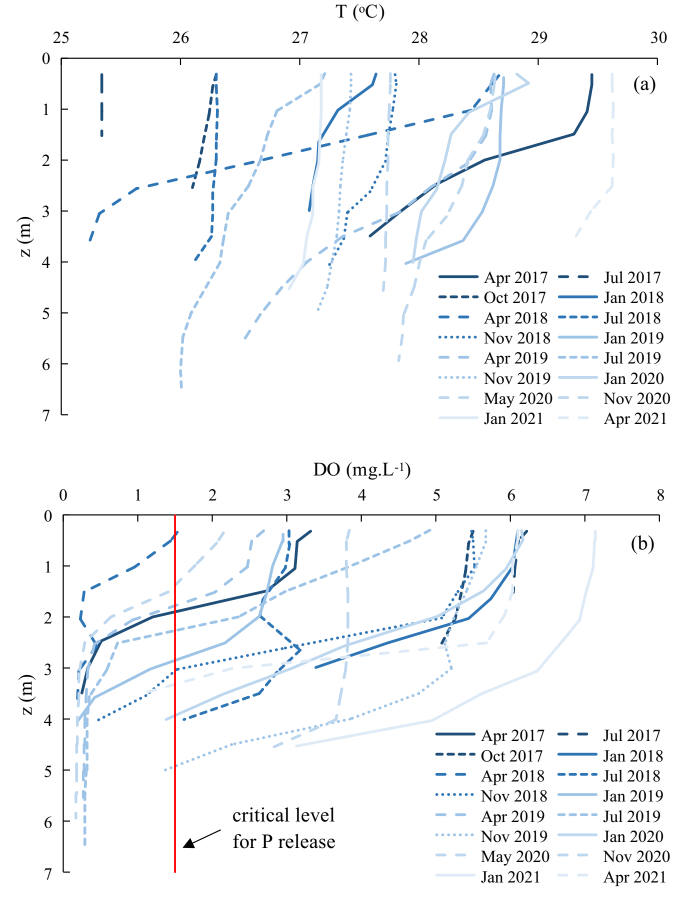

Fig. 5. Vertical profiles of water temperature (T) and dissolved oxygen concentration (DO), with indication of a critical DO level for P release (1.5 mg. L^{-1}). The total reservoir depth is 10 m.

3.5. Total phosphorus modeling

Phosphorus dynamics in the water column was modeled with Eq. (5) considering the three modeling schemes described in the Material and Methods section: i) Settling model; ii) Settling and release model with fixed sediment area (A_{sed}), where phosphorus is released to the water; iii) Settling and release model with variable sediment area (A_{sed}). Fig. 6 shows a comparison between model simulations and measurements of total phosphorus concentration in the water ($P_{T,out}$). It can be noticed that the model scheme proposed in this study, i.e., settling and release model with variable sediment area (A_{sed}), fitted well the measured data and presented the best performance, with NSE = 0.65 and RSR = 0.46. The settling and release model with fixed A_{sed} performed slightly worse, with NSE = 0.56 and RSR = 0.54. On the other hand, the simplest settling model provided the worst results, with NSE = 0.18 and RSR = 0.64. If one takes the NSE as a reference, the settling + release model with variable A_{sed} resulted in a "good" NSE value, while the settling + release model with fixed A_{sed} and the settling model resulted respectively in "satisfactory" and "not satisfactory" NSE values, according to Moriasi et al. (2015). Fig. 6 also reveals that higher P concentrations tend to occur during the wet periods, when the combined effects of external loadings from runoff events and internal loadings due to hypolimnetic hypoxia (see Fig. 5) and sediment P release favor phosphorus accumulation in the water column.

3.6. Total phosphorus loadings

Fig. 7 shows the simulated time series of the external and internal loads for current conditions of the 31-years-old São José I reservoir. A mean total load of 0.19 ton/month was obtained, resulting in a mean total areal load of

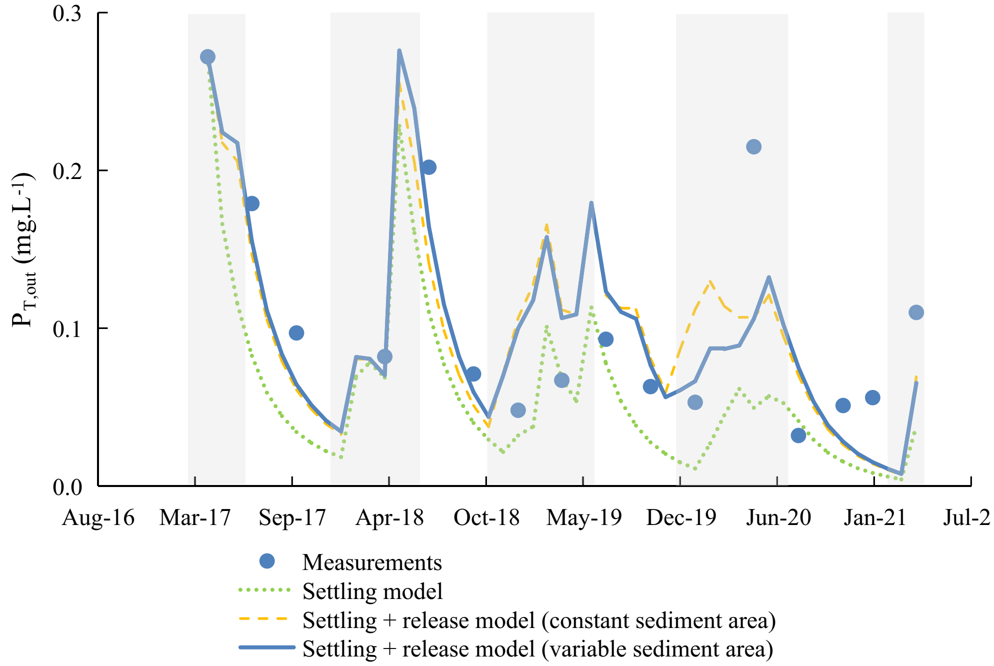

Fig. 6. Comparison between model simulations and measurements of total phosphorus concentration in the water column. Gray shades represent the wet periods.

1.37 g/(m^2.yr), which is within the ranges of 0.66–7.29 g/(m^2.yr) reported by Rocha and Lima Neto (2021b) for other reservoirs located in the same region. Additionally, the dry period load is about 11% of the total load, which is also within the ranges of 7–63% reported by Rocha and Lima Neto (2021b). The results also indicate that the internal load represents 44% of the total load, which highlights that it can be as high as the external load, corroborating previous studies available in the literature (Steinman et al., 2007; Nürnberg et al., 2012, 2013; Nikolai and Dzialowski, 2014). An unexpected result was that the internal load was predominant during the wet period (78% of the total internal load), when the reservoir level was higher and stratification/hypolimnetic hypoxia were more frequent. This explains why P concentration tends to be higher during the wet periods, as depicted in Fig. 6.

3.7. Impact of reservoir age on phosphorus dynamics

Fig. 8 presents the results of model simulations of total phosphorus for the current age (31 years) of São José I reservoir and a hypothetical age of 100 years, but keeping the hydrological and external load regimes the same for both cases. P_{FeAl} accumulation in the sediment leads to significant modification of the release rate of total phosphorus from the sediment to the water column, from $k_r = 165.0$ mg.m^{-2}.month^{-1} for current conditions to $k_r = 999.6$ mg.m^{-2}.month^{-1} for the hypothetical age of 100 years. This change increases P concentration in the water of about 3-fold (Fig. 8). Furthermore, for the age of 100 years, the average internal load potentially increases by about 6-fold, representing 83% of the total phosphorus load (see Fig. S3 in supplementary material), against 44% in current conditions (see Fig. 7).

3.8. Total phosphorus budget

Fig. S4 (supplementary material) shows the total P budgets considering both the 31-years-old and 100-years-old reservoir conditions. For the former case, the average external load ($Q_R.P_{T,in}$), settling rate ($-k_s.V.P_{T,out}$), release rate ($k_r.A_{sed}$) and output load $[-(Q_S + Q_W).P_{T,out}]$ correspond to respectively 0.105 ton/month (24%), −0.191 ton/month (44%), 0.084

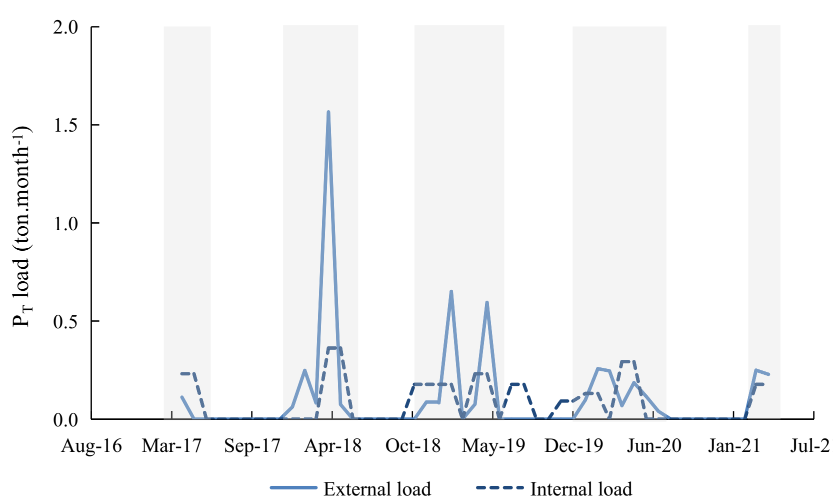

Fig. 7. Simulated time series of the external and internal phosphorus loads for current conditions (31-years-old reservoir). Gray shades represent the wet periods.

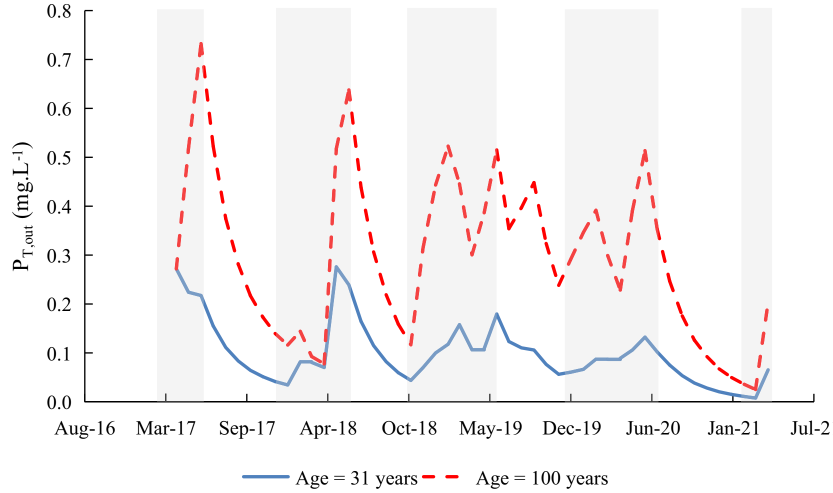

Fig. 8. Model simulations of total phosphorus for current (31-years-old) and hypothetical (100-years-old) age conditions of the São José I reservoir. Gray shades represent the wet periods.

ton/month (19%) and - 0.057 ton/month (13%). On the other hand, for the latter case, the external load, settling rate, release rate and output load correspond to respectively 0.105 ton/month (8%), −0.583 ton/month (42%), 0.507 ton/month (37%) and - 0.181 ton/month (13%). This indicates that keeping the external load but increasing the reservoir age from 31 to 100 years, the release rate increases as a result of higher k_r, while the settling rate and the output load also increase due to higher $P_{T,out}$. Note that in both cases the release rate is lower than the settling rate, but the ratio between these two variables increases from 44 to 87% as age increases from 31 to 100 years. It is also important to observe that while in the former case the reservoir is retaining phosphorus (output load < external load) at a rate of 0.048 ton/month, in the latter case, the reservoir is producing phosphorus (output load > external load) at a rate of 0.076 ton/month.

3.9. Implications for water management

The relevant internal P load indicated in this study brings new challenge to water management, particularly in tropical regions. For instance, water resources management in semiarid zones is based on the paradigm that the wet period is considered as most appropriate for water supply. However, higher P concentrations would also require higher investments in water quality management actions such as load reduction and/or water treatment improvement.

Furthermore, any efforts to control eutrophication by reducing the external P loads may be counteracted by the reservoir internal load. Considering the temporal accumulation of P_{FeAl} in the sediment, demonstrated in this and previous studies, late measures at the catchment scale to control phosphorus yield to the reservoir might not be effective to maintain the water quality at acceptable levels. Under such conditions, reservoir management techniques might be needed, such as hypolimnetic aeration/oxygenation to reduce phosphorus release (Bormans et al., 2016; Moura et al., 2020) or sediment recycling for soil fertilization (Braga et al., 2019; Lira et al., 2020).

3.10. Scientific contributions

The present paper contributes with new findings from field/laboratory studies on the behavior of the external/internal loads in tropical reservoirs as well as with important adaptations to the classical structure of P balance models. The scientific contributions of this study are summarized as following: (1) The external load can be described by an u-shaped relationship between inflow and phosphorus concentration; (2) The sedimentary concentration of P_{FeAl} increases along the reservoir and with reservoir age; (3) The internal load can be estimated from P_{FeAl} and variable heights of the anoxic layer; (4) The proposed model, which accounted for the variability of reservoir volume, an innovative formulation for the internal load and a different parameterization for the settling rate, significantly improved the prediction of phosphorus concentration in the water; (5) The internal load prevailed in the wet season and was slightly lower than the external load; (6) The internal load would increase by about 6-fold if the reservoir were 70 years older; and (7) The reservoir can act as a sink or source of phosphorus, depending on its age.

4. Conclusions

Modeling of phosphorus dynamics in the tropical 31-years-old São José I reservoir (7.7 10^6 m^3 storage capacity), supported by hydrological, meteorological and water quality data, as well as analyses of phosphorus partitioning of the bed sediment, enabled us to estimate phosphorus loadings and to draw important conclusions for water management in tropical regions.

The external phosphorus load was estimated from a relationship adjusted between reservoir inflow and total phosphorus concentration measurements, revealing an u-shaped behavior which is consistent for non-point source dominated catchments. The fitted relationship was also consistent with the data reported in the literature for other reservoirs located in the same region and with similar water storage capacities.

Sedimentary phosphorus linked to Fe and Al (P_{FeAl}) increases from the riverine to the lacustrine zone and according to reservoir age. This finding is particularly important for water quality assessment, as previous studies have proven that P_{FeAl} is responsible for phosphorus exchange from the sediment to the water column under anoxic conditions, representing a lake internal source.

The internal phosphorus load, i.e., the portion originating from the reservoir bed sediment, accounts for 44% of the total load in current conditions (31-years-old reservoir), therefore is only slightly lower than the external source. It prevailed during the wet season, when reservoir stratification and low dissolved oxygen levels were more notable, resulting in higher phosphorus concentration in the water column due to the combined effects of external and internal loadings. This implies that while the rainy events promote higher volumes for water supply, water quality can be compromised due to the additional impact of phosphorus release from bed sediments.

For hypothetical conditions of reservoir age (100-years-old reservoir), the internal load reaches 83% of the total and the reservoir becomes a source of phosphorus to the downstream river. This pattern indicates that, after long periods of high external loads with phosphorus accumulation in the sediment, measures to control external phosphorus yield to reservoirs might not be sufficient to control eutrophication. In such cases, reservoir management techniques might be needed to maintain phosphorus concentration in the water at acceptable levels.

The complete-mix phosphorus budget model adopting first- and zero-order kinetics to simulate phosphorus settling and release from the sediment layer, respectively, was able to reproduce the phosphorus temporal dynamics in the reservoir. The adaptations proposed in this study to the original Vollenweider's model in order to account for water level (volume) changes, the internal load using an innovative formulation, and a different parameterization for the settling process, improved the model capacity to estimate phosphorus concentration, from "not satisfactory" to "good". The modeling scheme proposed in this study represents a useful tool to water management in tropical regions, supporting the assessment of phosphorus dynamics in reservoirs.

CRediT authorship contribution statement

Iran E. Lima Neto: Conceptualization, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Supervision, Writing – original draft, Writing – review & editing. **Pedro H.A. Medeiros:** Conceptualization, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Writing – review & editing. **Alexandre C. Costa:** Conceptualization, Formal analysis, Investigation, Methodology, Writing – review & editing. **Mario C. Wiegand:** Data curation, Formal analysis, Investigation. **Antônio Ricardo M. Barros:** Data curation, Formal analysis, Investigation. **Mário U.G. Barros:** Data curation, Formal analysis, Resources.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.152875.

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